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The electron capture decay of ^{228}Pa[1]

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With 1 figure in the text

ABSTRACT

The radiations of ^{228}Pa were investigated with a double-focusing beta-ray spectrometer and with a scintillation counter. 43 conversion lines were detected, which could be ascribed to 18 gamma-transitions. The energies of most of these transitions agree fairly well with those found in the decay of $MsTh_2$. Multipolarities were assigned to the different transitions. A decay scheme is suggested based on the γ–γ coincidence measurements of Box and Klaiber, comprising a rotational level system at 57.48 and 186.1 keV and perhaps also at 1070 and 1125 keV.

1. Introduction

According to Meinke *et al.* [2] ^{228}Pa decays partially by α-particle emission, with a transition energy of 6.20 MeV in 2 % of the cases; 98 % of the nuclei decay by electron capture to excited levels in ^{228}Th. The total half-life is 22 ± 1 hr. No measurements of the γ-transitions following the E.C.-decay have been reported. Since the excited levels of ^{228}Th are also populated by the negatron decay of $MsTh_2$ (^{228}Ac), more is known about the excited levels of ^{228}Th. A detailed magnetic analysis of $MsTh_2$ was made by Black [3] in 1924, in which he used a semicircular focusing spectrograph. According to Hollander *et al.* [4], Jenkins and O'Kelley have found 7 γ-rays and 3 β-components in $MsTh_2$. Campbell *et al.* [5] and Kyles *et al.* [6] investigated the electron spectrum with a permanent magnet β-ray spectrometer. They found 6 partial β-ray spectra and 38 conversion lines, corresponding to 17 γ-ray energies. Their experiments were supplemented by those of Brodie [7] who used a lens spectrometer with post-focusing acceleration, which enabled him to measure down to 4 keV. From these data Kyles *et al.* suggested a decay scheme in which the highest energy beta-transition populates an excited state of ^{228}Th at 57.5 keV.

Recently Box and Klaiber [8] published their results of single and coincidence γ-ray measurements in $MsTh_2$ with two scintillation spectrometers. In addition to some γ-transitions reported by Kyles *et al.* they found new transitions of 155, 220, 278, and 790 keV. From their coincidence measurements they proposed a level scheme of ^{228}Th, that deviated from Kyles' decay scheme.

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Considering that an investigation of the E.C.-decay of ^{228}Pa might give some additional information about the excited levels in ^{228}Th, we started on a measurement of the electron spectrum of this isotope. In the course of our investigation the results of Passell [9] on the E.C.-decay of ^{228}Pa were reported. The activity was obtained by bombardment of thorium with 115 MeV protons. Four electron lines could be positively assigned to ^{228}Pa as the L_{II} and L_{III} conversion lines of the γ-rays of 57.8 ± 1 and 130 ± 2 keV.

2. Experimental work

The ^{228}Pa activity was obtained by the Th$(p, 5n)$ reaction on thorium powder in the Uppsala synchrocyclotron. We bombarded with several proton energies between 50 and 80 MeV in order to investigate the optimal conditions for the Th $(p, 5n)$ reaction. The protactinium activities were separated from the target material and most of the fission activities by means of the solvent extractions with diisopropyl ketone and thenoyl-trifluoro acetone according to the procedure given by Meinke [10]. Although this procedure gave a good purification factor, it was difficult to get rid of all the fission activities (e.g. Zr, Nb) without appreciable loss of the protactinium activities. Other interfering activities were those of ^{229}Pa, ^{230}Pa and ^{232}Pa.

The preliminary measurements were performed in the 30.0 cm double-focusing spectrometer at Amsterdam [11]. For that reason the target material was transported by plane from Stockholm to Amsterdam so that we could start with the chemical separation five hours after the irradiation. The transmission of the spectrometer was 1.2 % and the resolution 0.9 %, the source width being 0.6 cm. The activity was put onto a LC 600 foil wetted with insulin. The beta counter had a cut-off energy of 35 keV.

The final experiments were done in the 18.5 cm spectrometer at Uppsala [12], adjusted to a transmission of 0.8 % and a resolution of 0.65 %. The activity was evaporated under vacuum onto thin aluminium foils. The electron spectrum could be measured down to 18 keV by using counter windows of rubber hydrochloride films. The spectrum was scanned several times with the automatic recording arrangement in order to get a rough estimation of the energy and the half-life of the conversion lines. Afterwards the conversion lines were measured manually.

The combined results of the measurements done in Amsterdam and Uppsala are shown in Table 1. In column 3 the energy values of the electron lines found in our measurements are given. From the shell assignments (column 6) follow the corresponding energies of the γ-rays as given in column 7. The half-lives of these lines were measured, whenever possible, in order to be sure that they did not belong to other activities. Taking 22 hours as the right value of the half-life, the relative intensities of the various lines were estimated (column 4). By way of comparison the energies of the conversion lines, observed by Black and by Kyles *et al.* in MsTh$_2$ are also given in the columns 1 and 2 respectively.

Three electron lines have been ascribed to Auger lines, while three conversion lines at 624, 648 and 1230 keV, not listed in Table 1, could not be assigned to certain transitions.

No separate samples were prepared for the γ-ray measurements. Old samples of one day or older, which were used in the beta spectrometer, were employed for this purpose. The NaI crystal was 2 cm thick with a diameter of 2 cm and coupled to a Dumont 6292 multiplier. The average energies of the photo peaks and the

Table 1. Conversion lines of ^{228}Pa and MsTh$_2$.

1	2	3	4	5	6	7
E_{e^-} (keV)		This investigation				
Black	Kyles*	E_{e^-} (keV)	I_{e^-}	$T_{1/2}$ (hr)	Shell	E_γ (keV)
—	—	19.1			K	128.7
—	36.34	—				
37.5	36.91	37.84	20	18	L_{II}	57.51
40.9	40.33	41.14	20		L_{III}	57.42
—	(43.14)	—				
—	(47.28)	46.2			K	155.8
—	(50.47)	—				
52.2	52.16	52.59	8	26	M_{II}	57.40
—	—	53.49			(M_{III})	57.52
55.5	55.41	56.39	3.8	20	N_{II}	57.54
58.2	58.19	—				
61.9	61.91	61.4			L_{III}	77.7
66.9	66.97	68.0			$K \rightarrow L_{\mathrm{I}}L_{\mathrm{I}}$	
	71.03	72.6			M_{II} $K \rightarrow L_{\mathrm{I}}L_{\mathrm{III}}$	77.4
73.4	73.76	74.9	2.5		K	184.5
77.7	77.98	77.3			L_{II}	97.0
—	81.49	79.9			L_{III}	96.2
—	—	84.7			$K \rightarrow L_{\mathrm{I}}M_{\mathrm{I}}$	
92.2	92.66	92.0			M_{II}	96.8
—	97.04	95.4			N_{II}	96.6
107.4	108.1	108.89	4.8	21 ± 2	L_{II}	128.56
111.01	111.3	112.30	3.2	19 ± 2	L_{III}	128.58
	(114.8)	114.1			K	223.7
122.4	122.2	123.85	2.2	22 ± 2	M_{II}	128.66
125.8	125.8	127.59	0.6	21 ± 2	N_{II}	128.74
131.6	(131.6)	—				
138.3	(140.3)	—				
—	(145.8)	—				
—	157.0	—				
161.6	164.9	164.0	0.4	25	L_{I}	184.5
175.3	178.7	173.3	1.0	22 ± 2	K	282.9
206.6	(202.4)	203.8	0.40	22	L_{I}	224.3
—	212.0	218.2	0.35		K	327.8
225.7	225.9	227.5	—		K	337.1
—	—	262.5	0.32	27	L_{I}	283.0
—	—	278.6	—		M_{I}	283.8
295.4	300.5	300.7	0.40	20	K	410.3
313.7	315.9	309.2	0.25		L_{I}	329.7
—	—	336.4	0.40		K	446.0
348.1	348.6	354.8	0.90	23	K	464.4
—	—	389.9	0.25		L_{I}	410.4
—	—	407.3	0.10		M	412.5
—	—	424.3	0.06	20	L_{I}	444.8
437.0	437.0	445.0	0.40	22	L_{I}	465.5
451.9	451.9	461.8	0.20	23	M	467.0
—	—	683	—		K	793
—	—	773	0.15		L_{I}	793.5

* Lines between parentheses were not assigned to any transition.

Table 1 (cont.).

1	2	3	4	5	6	7
E_{e^-} (keV)		This investigation				
Black	Kyles	E_{e^-} (keV)	I_{e^-}	$T_{1/2}$ (hr)	Shell	E_γ (keV)
796.1	796.4	803.5	0.50	23	K	913
851.8	858.5	860	0.50	23	K	969.6
889.1	888.6	894	0.12		L_I	914.5
—	925.1	—	—		—	
940.4	940.4	947	0.14		L_I	967.5
—	985.1	985	—		K	1095
1483	1477	1462	—		K	1572
1522	1530	—	—		—	—

Table 2. Gamma-rays of ^{228}Pa.

E_ph (MeV)	I_rel
1.58	6
0.95	40
0.46	20
0.41	8
0.33	10
0.28	—
0.23	18
0.184	4
0.090	100

relative intensities of these γ-rays corrected for the photon efficiencies of the crystal, are shown in Table 2.

3. Results

The mean γ-ray energies, calculated from the energies of the conversion lines, are tabulated in Table 3, where γ-ray energies in the decay of $\mathrm{MsTh_2}$, reported by Kyles *et al.* and by Box and Klaiber, are incorporated for comparison. Most of the transitions observed in $\mathrm{MsTh_2}$ have also been found in the decay of ^{228}Pa. It is surprising, however, that the energies found by Kyles *et al.* and by us differ to a considerable degree. The conversion lines belonging to the reported transitions of 113, 1035 and 1640 keV were not observed by us. Weak conversion lines of energies 46.2, 114.1 and 203.8 keV, observed by Kyles *et al.* but not assigned to any transitions, are ascribed to the transitions of 156 and 224 keV in agreement with the results of Box and Klaiber. A very weak electron line at 227.5 keV is assumed to be the K-conversion peak of the reported 336 keV transition. Its L-conversion line was not found. The reason might be that it coincides with the 320.7 keV line of ^{233}Pa [1]. Weak conversion lines are ascribed to γ-rays of energies 328.5 and 445.4 keV, although these transitions have not been reported in the decay of $\mathrm{MsTh_2}$.

Table 3. Gamma transitions of ^{228}Pa.

This investigation				Kyles *et al.*	Box and Klaiber
γ-ray No.	E_γ (keV)	K/L	Multipole	E_γ (keV)	E_γ (keV)
1	57.48		$E2$	56.8	
2	77.7		$(E2+M1)$	78.1	
3	96.6		$(E1)$	97.8	98
				113.0	
4	128.64		$E2$	127.5	127
5	156		$(E1)$		155
				179.0	
6	184.5	6.0 ± 1.0	$M1$	184.2	
7	224.0	—	—	232.2	220
8	283.2	3.2 ± 0.6	$E2+M1$		278
9	328.5	—	—		
10	337.0	—	—	336.0	336
11	411.0	2.5 ± 0.5	$E2\,(+M1)$	410.1	410
12	445.4	7 ± 2	$(M1)$		
13	465.6	2.2 ± 0.5	$E2$	457.6	458
14	793	—	—		790
15	914	4 ± 1	$E2\,(+M1)$	907.1	935
16	968	3.5 ± 1	$E2\,(+M1)$	964.5	
				1035	
17	1095	—	—	1095	
18	1572	—	—	1587	1587
				1640	

In Table 3 the K/L conversion ratios observed in our measurements are also given. Since many of the electron lines are rather weak, most of these ratios have to be regarded as rough estimates. Some of the values do not agree with those reported by Kyles *et al*. Considering that the background in our measurements is much smaller, because of the absence of a continuous spectrum belonging to ^{228}Pa, we expect our values to be more reliable. Multipolarities are determined by comparison of the observed K/L ratios with the theoretical calculations of Rose [13]. The 57.48 and 128.6 keV transitions are predominantly converted in the L_{II} and L_{III} subshells. This seems to indicate that both transitions should be $E2$. The K/L ratio of the latter transition, which is 0.023 according to Brodie, also indicates this multipolarity.

4. Decay scheme

Because ^{228}Th, the daughter of ^{228}Ac, is an even–even nucleus, its ground state should be $0+$. The rotational excited states predicted by the Bohr-Mottelson model [14] should be de-excited by a cascade of $E2$ transitions. For this reason we suppose the 57.48 and 128.6 keV transitions to de-excite the levels at 57.48 keV $(2+)$ and at 186.1 keV $(4+)$. The $E(4+)/E(2+)$ ratio then amounts to 3.24 in agreement with the Bohr-Mottelson model (see also reference 15). The assumption of $2+$ and $4+$ excited levels at 57.48 and 186.1 keV is supported by the result of α-decay measurement in ^{232}U by Asaro and Perlman [16]. They report three α-groups leading to the ground state and excited states at 57 and 186 keV in ^{228}Th. The partial α-group intensities (68 %, 32 % and 0.32 %) agree with the α-group intensities of other even-even nuclides, which decay to 0+, 2+ and 4+ states of their daughter nuclides.

Since a direct transition from the 4+ state is not probable, the 184.5 keV transition is sure not to be the crossover transition. We believe the observed difference between the sum of the energies of the two γ-rays of 57.48 and 128.6 keV and the 184.5 keV γ-ray energy to be real. Kyles *et al.*, however, assumed the 184.2 keV γ-ray to de-excite a 1+ state at 184.2 keV. For this reason they supposed the 127.5 keV transition to be a mixture of $E2$ and $M1$.

Two different level schemes of ^{228}Th were proposed by Kyles *et al.* and by Box and Klaiber. We assumed the latter scheme, because the γ–γ coincidence measurements of Box and Klaiber give more reliable results about the position of cascading γ-rays. However, we use our mean energies of the γ-transitions, because we believe our beta spectrometric measurements to be more accurate. Our only alteration in the level scheme proposed by Box and Klaiber is, that we assumed a new excited level at 1720 keV, which should be de-excited by the 77.7 and 184.5 keV γ-ray transitions. The observed γ-rays with energies of 328.5, 445.4 and 1095 keV do not, however, fit into the level scheme (Fig. 1).

In a first report [1] we discarded the presence of an excited level at 337 keV and assumed an excited level to exist at 790 keV, which should be populated by the 328.5 and 283.2 keV transitions from the levels at 1125 and 1070 keV. Recently, however, Stephens *et al.* [17] observed an excited 1-state of ^{228}Th at 330 keV, in their study of the α-decay of ^{232}U. This might support Box' assumption of a level at 337 keV. This level might be de-excited by $E1$ transitions leading to the ground state and to the first rotational 2+ state. The K/L conversion ratio of the 283.2 keV transition, however, does not indicate an $E1$ radiation, but is more compatible with a mixture of $E2$ and $M1$. The excited level at 156 keV also decays to the ground state and the 2+ state. It is therefore quite possible that this level is a 1− excited state. The transitions of 96.6 and 156 keV might then be $E1$ radiations, which is not in contradiction with the experimental data. It is, however, strange that this level is not found to be populated by alpha-decay of ^{232}U. It seems therefore worth-while to remeasure more accurately the K/L conversion ratios and the α_K-values of these transitions.

In order to make this level scheme consistent with the β-transitions found by Kyles *et al.*, we have to assume the following alterations. Kyles *et al.* have found 6 partial β-components with maximum energies of 2.18 (10 %), 1.85 (9 %), 1.70 (7 %), 1.11 (53 %), 0.64 (8 %) and 0.45 (13 %) MeV. Moreover, they assumed that the first component does not decay to the ground state of ^{228}Th, but to the 57.5 keV excited level. According to their coincidence measurements this should be an isomeric level with a half life of about 10 msec. Assuming that the high energy β-component decays to the ground state, we may obtain from our level scheme β-transitions of 1.77, 1.11, 0.645 and 0.46 MeV. The last three transitions agree rather well with the values obtained by Kyles *et al.* Instead of the two transitions of 1.85 and 1.70 MeV, we assume only one transition of 1.77 MeV. This is not inconsistent with the Fermi-Kurie analysis of Kyles' measurements. If the assumption is maintained that the 2.18 MeV β-component decays to the 57.5 keV excited level, the other β-components do not fit well, with the energies of the excited levels in the proposed level scheme.

Since the 914 keV and/or 968 keV transitions might be a mixture of $M1$ and $E2$, the levels at 1070 and/or 1125 keV should have odd parity. This is also the case with the levels at 1535 and at 1720 keV. The β-decay of MsTh$_2$ indicates first-forbidden

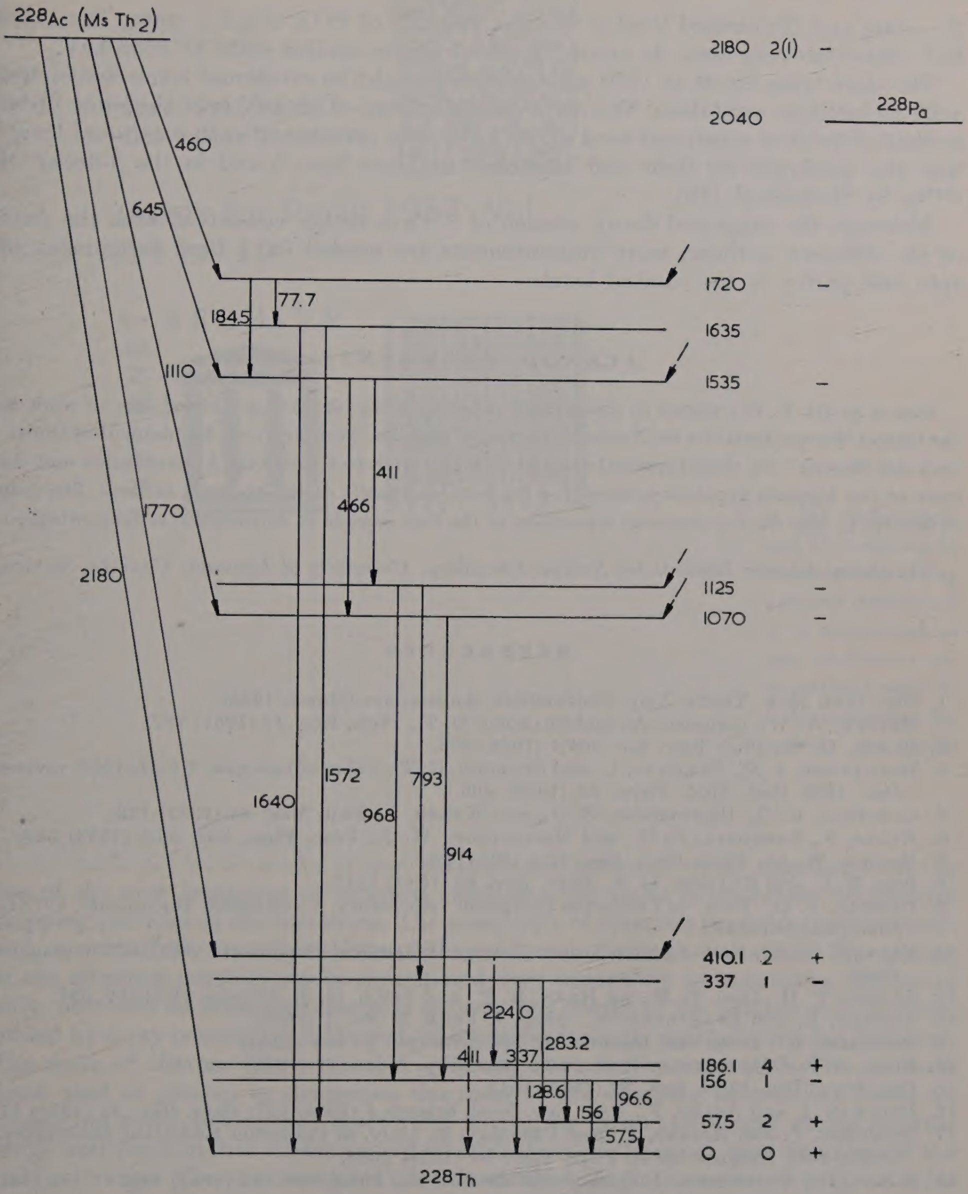

Fig. 1. Decay scheme of ^{228}Pa and MsTh$_2$.

transitions for the higher energy components, while the lower energy components might be allowed. This indicates that the ground state parity of ^{228}Ac should be odd, not in contradiction with the predictions of the single particle model. The spin might be 2 or 1. In our decay scheme we assume the ground state of ^{228}Ac to be in the

2 − state and the excited level in ^{228}Th at 410 keV of ^{228}Th in a 2 + state. The 411.0 keV transition may then de-excite this level or the excited state at 1535 keV.

The close-lying levels at 1070 and 1125 keV might be rotational levels associated with an intrinsic excitation. The difference in energy of 55 keV is of the same order as that of the first rotational level (57.48 keV). The presence of such rotational levels was also predicted by Bohr and Mottelson and has been found in the β-decay of ^{238}Np by Slätis *et al.* [18].

Although the suggested decay scheme of ^{228}Pa is rather consistent with the data of the different authors, more measurements are needed for a final assignment of spin and parity to the excited levels.

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